

Charge density wave transport in submicron antidot arrays in NbSe₃

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We demonstrate for the first time that a periodic array of submicrometer holes (antidots) can be patterned into thin single NbSe₃ crystals. We report on the study of charge density wave (CDW) transport of the network of mesoscopic units between antidots. Size of the elementary unit can be as small as 0.5 μm along the chain axis and 0.2 μm×0.3 μm in cross section. We observe size effects for Ohmic residual resistance and in CDW transport current-voltage characteristics in submicronic networks.

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I. INTRODUCTION

There is a considerable interest in the study of mesoscopic CDW structures the characteristic size of which is comparable with the coherence length for the phase or the amplitude of the CDW order parameter. From these studies it is expected to obtain new informations about the intrinsic coherence of the CDW conduction¹. Recent experimental and theoretical investigations have demonstrated the possibility of the observation of quantum interference effects in heterostructures^{2,3} and nanostructures⁴ in CDW materials. Progress in mesoscopic CDW studies is foreseeable with the recent development of epitaxial thin film technology⁵ combined with electron lithography technique. In the present publication, we report on CDW transport in submicron structures patterned on thin single crystal of the CDW quasi one-dimensional (Q1D) compound NbSe₃.

II. EXPERIMENTAL

The idea of the experiment was to carry out the realization of a mesoscopic structure in which CDW motion is localized in an array of small cells with a submicron size, each cell being electrically connected to the neighbouring ones by transverse (to the chains) conduction. We show in Fig. 1 a schematic example of such a structure. The chain direction of Q1D NbSe₃ is along x axis. A triangular lattice of holes (antidots) is made in such a way that the projection of holes along the y direction overlaps, that provides small units of CDW conduction between neighbouring holes. The individual cell is shown in Fig. 1 as a shaded rectangle.

We selected the most perfect thin NbSe₃ crystals with a thickness, d , less than 1 μm and a relatively large width w ($w > 20$ μm). The width is along the c crystallographic axis. The NbSe₃ crystals were first glued onto a silicon or sapphire substrate using a fluid resist. After baking 10 minutes at 130°C the residues of resist were removed by a soft oxygen plasma etching. The crystal were then thinned using SF₆ reactive ion etching down to about 0.5 μm thickness. A 25 nm thin layer of silicon is then deposited by e-beam evaporation to protect the surface of the crystal and insure charge evacuation from the sapphire substrate during the subsequent e-beam lithography.

The desired holes have high aspect ratio: the hole size is 0.4×0.6 μm, wall thickness is about 0.2 micron and crystal thickness is 0.5 μm. One therefore needs a sharp metallic mask. We first spin on a PMMA layer of nominal thickness 0.5 μm and bake it 160°C during 10 minutes. This PMMA layer is then patterned by direct writing with the electron beam of a scanning electron microscope and developed. We prepare a 50 nm aluminum mask by angle evaporation onto the surface of the patterned PMMA layer.

The etching of NbSe₃ is then achieved in a SF₆ plasma with the aluminum film. We have checked optically that for most of the samples the depth of etching coincides with the thickness of the crystal itself. We then remove the aluminum mask by wet etching and the PMMA layer in an oxygen plasma.

We have measured two NbSe₃ samples with triangular lattice of holes, keeping nearly the same the ratio between the antidot lattice periodicity and the average diameter of the holes. The scanning electron micrographs in the patterned parts are shown in Fig. 2. The triangular lattice of antidots for sample # 1 (deposited on a silicon substrate) as shown in Fig. 2a has a periodicity $S_b = 3$ μm along the b axis and $S_c = 3.7$ μm along the c axis. The hole cross section nearly circular is 1.4 μm × 1.5 μm. For sample # 2 (deposited on a sapphire substrate) the periodicity of the lattice

of antidots is $S_b = S_c = 0.6 \mu\text{m}$ with a rectangular hole cross section with $D_b = 0.3 \mu\text{m}$ along the b axis and $D_c = 0.4 \mu\text{m}$ along the c axis (see Fig. 2b).

Four gold contact pads are then patterned in a second e-beam lithographic step using lift-off technique. External leads are attached to the gold strips using indium contacts. The contact geometry is chosen to allow measurements on adjacent patterned and non-patterned zones (typical length: 100 μm) of the same crystal as drawn in Fig. 3. The microhole structure covers the whole width of the crystal.

The electrical resistance and the differential current voltage characteristics were measured with a 40 Hz phase sensitive bridge using the inner probes 1-2 and 3-4 as voltage probes to compare the properties of the patterned and non-patterned segments.

All the sample parameters are indicated in Table 1.

III. RESULTS

The processing treatment yields some changes in the properties of the NbSe₃ samples. In Fig. 4a we show the temperature dependence of a NbSe₃ sample before processing and after processing (essentially the baking of the PMMA layer at 160°C). The consequences of processing are an increase of both Peierls transition temperatures ($T_{P_1} = 145$ K and $T_{P_2} = 59$ K for pure NbSe₃ crystals), an increase of the resistance at both maxima in the $R(T)$ dependence and a larger residual resistance at low temperatures. However the sample still conserves all the properties of a relatively good quality NbSe₃ crystal. After processing but without patterning the resistance ratio $R(290\text{ K})/R(4.2\text{ K})$ is in range of $\sim 25\text{-}40$ and the threshold electric field for CDW depinning at 50 K is $E_T \sim 100$ mV/cm.

Patterning the antidot array strongly affects the resistance of the NbSe₃ sample. In what follows each NbSe₃ sample measured will have been processed on its total surface but only a part is patterned (see Fig. 3). The sheet resistance of the patterned part increases considerably, for instance by a factor 6.5 at room temperature for sample # 2; however the Peierls transition temperatures are not changed with respect to the unpatterned part as shown in Fig. 4b. The effect of patterning is clearly seen in the temperature dependence of the ratio between the resistance of the patterned part and the resistance of the unpatterned part. Prominent features appear near both Peierls transition temperatures and at low temperatures below 50 K. The decrease of the resistance of the patterned part below 50 K is largely suppressed for sample # 2 with the shorter antidot lattice periodicity. Thus, the resistance ratio between room temperature and helium temperature is 10 for sample # 1 and 4.8 for sample # 2 (to be compared to 25-40 for the unpatterned zone).

Fig. 5 shows the variation of the differential resistance $\frac{dV}{dI}$ as a function of the applied current for both non-patterned (Fig. 5a) and patterned (Fig. 5b) parts of sample # 2. CDW motion occurs above a threshold electric field E_T defined as $E_T = \frac{V_T}{L} = \frac{RI_T}{L}$ with R the Ohmic resistance, L the distance between electrodes and I_T the critical current at which non linearity in the $(I - V)$ characteristics appears. It can be noted that I_T for the lower CDW is nearly identical for the patterned and unpatterned parts as shown in Fig. 6.

IV. ANALYSIS AND DISCUSSION

We will now describe a simple model in order to explain the properties of submicronic structures patterned on a single NbSe₃ crystal. We consider that: 1) transport along chains occurs only in the short cells located between neighbouring holes (shaded part in Fig. 1) and 2) transport from one cell to the adjacent one requires charge motion perpendicularly to the chains. One can imagine an equivalent scheme which consists in an electrical network with two types of resistances: one, R , for transport along the chains, the other, r , for transport across the chains as drawn in Fig. 7.

A. Ohmic behaviour

The ratio between the sheet resistance of the patterned part, R , and that of the unpatterned one, R° , $G = \frac{R}{R^\circ}$ can be expressed as a linear function of r/R , which is proportional to the resistivity anisotropy ρ_c/ρ_b (ρ_c : resistivity along the width of the sample, ρ_b : resistivity along the chain axis). Thus, $G = A + B(\rho_c/\rho_b)$ where A and B are coefficients depending on the geometry of the network. In the case where $A \sim 1$ and $\rho_c/\rho_b \gg 1$ which is appropriate in the present case, one can write:

$$G = B \frac{\rho_c}{\rho_b} \quad (1)$$

For sample # 2, we find $G = 6.5$ at room temperature. Using the value of $\rho_c/\rho_b \sim 15$ measured at room temperature in NbSe_3^6 , one obtains $B = 0.43$. The justification for the validity of our conduction model is shown in Fig. 8 where we have drawn the temperature dependence of $\frac{G(T)}{B}$ with $B = 0.43$ and that of the resistivity anisotropy ρ_c/ρ_b measured in⁶ using the Montgomery technique. A good agreement between the temperature dependence of both quantities is found in the full temperature range above $T \simeq 55$ K (remember that T_{p1} and T_{p2} are larger in our processed samples). At lower temperatures, Eq. 1 is unvalid, because the additional contribution to the resistance due to the patterned structure. We consider that the origin of this additional contribution comes from the additional scattering in the mesoscopic units, when the mean free path, ℓ , becomes comparable at low temperatures to the length of the unit along the chains. Using the data from⁷ for the carrier mobility along chains, we estimate that ℓ is $\sim 0.3 \mu\text{m}$ at $T = 40$ K. Below this temperature, the increase of ℓ becomes limited by the antidot periodicity. That can explain the decrease of the residual resistance ratio from 10 (sample # 1) to 4.8 (sample # 2) when the antidot lattice periodicity along the b axis is reduced from $S_b=3\mu\text{m}$ (sample # 1) to $0.6 \mu\text{m}$ (sample # 2).

B. Threshold field for CDW depinning

We consider now the CDW depinning in a network structure. The threshold we have defined in part 3 corresponds to a value averaged over the whole structure. Let it to be noted as : $\langle E_T \rangle = \frac{R}{L} \langle I_T \rangle$.

However, it is also possible to define a local threshold field, noted as E_t , in an elementary cell : the shaded rectangle for the patterned part as shown in Fig. 1 with dimensions Λ along the b axis and Λ' along the c axis. If the elementary cell extends half the distance between units along b axis as represented in Fig. 1, then $\Lambda = S_b$ and $\Lambda' = S_c - D_c$. For the unpatterned part, the elementary cell is larger with dimensions S_b along the b axis and S_c along the c axis. E_t is connected with the local current I_t , along the chains in that elementary cell such as $I_t = \sigma_b E_t S$ where σ_b is the linear conductivity along the chains and S the cross section of the sample. The ratio between E_t in the patterned part and the threshold field of the non-patterned part, that one notes as E_t^0 , can be expressed as follows:

$$\frac{E_t}{E_t^0} = \frac{I_t}{I_t^0} \frac{\sigma_b^0}{\sigma_b} \frac{S_c}{S_c - D_c}$$

One then makes the reasonable assumption that the local $\frac{E_t}{E_t^0}$ is the same than the average value for the whole sample $\langle \frac{E_t}{E_t^0} \rangle$.

For sample # 2 we have shown the temperature dependence of $\langle I_T \rangle$ and $\langle I_T^0 \rangle$ in Fig. 6. The temperature variation of $\frac{\langle I_T \rangle}{\langle I_T^0 \rangle}$ for sample # 1 and # 2 are drawn in Fig. 9.

For sample # 1, with a larger cell size ($D_c = 1.4 \mu\text{m}$, $D_b = 1.5 \mu\text{m}$), we find that $\langle I_t/I_T^0 \rangle \sim 0.7$ with no significant temperature dependence. For this sample, $\frac{S_c}{S_c - D_c} = 1.6$; one neglects any size effect for σ_b such as $\sigma_b^0/\sigma_b \sim 1$. Then $E_t/E_t^0 \sim 1.1$ not far from the value of 1.

For sample # 2, with $D_b = 0.3 \mu\text{m}$ and $D_c = 0.4 \mu\text{m}$, we measure $\langle I_T/I_T^0 \rangle = 1.2$ (Fig. 9). With $\frac{S_c}{S_c - D_c} = 3$, one gets $E_t/E_t^0 \approx 4$, demonstrating that there is a well pronounced size effect for the local E_t in a cell with submicron size. Similar size effects have been previously observed in TaS_3 single crystals with a very small cross section⁸ and a short length⁹.

C. Current conversion

One of the aims of our study is the conduction due to CDW motion in a submicron unit. The process of conversion of the normal carrier current into a CDW current is still not well understood. In several theoretical models the conversion process is accompanied by auto-localization of carriers¹⁰, formation of phase-slippage centers¹¹, dynamical amplitude solitons¹², tunneling between solitons under the barrier¹³.

Phase slippage at current injection electrodes has resulted from experiments in which the variation of the threshold voltage was studied as a function of the distance between electrodes. It was found¹⁴, that :

$$V_T = E_p L + V_0$$

The first term corresponds to the bulk pinning due to impurities and the extra term V_0 was interpreted as the potential necessary for the nucleation of a CDW dislocation loop for conversion of normal current into the CDW

condensate. Typically for NbSe_3 V_0 is in the range of 0.2-0.5 mV at $T = 40 \text{ K}$ ¹⁴.

One can estimate the threshold voltage in our elementary cell of the patterned zone. The local critical current is the total critical current $\langle I_T \rangle$ divided by the number of channels along c axis. The resistance of the elementary cell can be calculated according to its dimensions and the conductivity at the given temperature. Thus, one estimate that at 40~50 K, the threshold voltage in the elementary cell is in the range of 50~100 μV , much less than V_0 . One are led to conclude that in our network geometry with submicronic dimensions where the current is fed to the elementary cell without any interface with a normal metal, there is no vortex dislocation loop generated.

The other models^{10,12,13} estimate the conversion length, L_{conv} , to be of the order of $\hbar v_F / \Delta$ (Δ : the CDW gap). An estimate of L_{conv} for NbSe_3 is $\sim 20 \text{ nm}$. One can expect to determine a value of the conversion length from experiments on short samples with a length Λ comparable with L_{conv} . In such a short sample only a part of the length of the sample, $\Lambda - 2L_{\text{conv}}$, contributes to the non-linear conduction. A comparison with a macroscopic sample with the same thickness and the same ratio length/width can be made. It is then expected that in small samples the maximum contribution to the non-linear conduction will be at least $\frac{\Lambda - 2L_{\text{conv}}}{\Lambda}$ smaller.

We have measured the non-linear properties in the current regime where the CDW conduction saturates i.e. $I \approx 10\langle I_T \rangle$ for both patterned and unpatterned parts of sample # 2. We obtain that the CDW conductivity in an individual cell, σ_{CDW} , is always smaller in the patterned part of the sample with respect to the value in the non-patterned part of the same sample, σ_{CDW}^0 . Fig. 10a shows the temperature dependence of $\gamma = \sigma_{\text{CDW}} / \sigma_{\text{CDW}}^0$. Making the suggestion that this ratio γ is a measurement of $\frac{\Lambda - 2L_{\text{conv}}}{\Lambda}$, one can estimate L_{conv} as equal to $\frac{\Lambda(1-\gamma)}{2}$. That yields a value of L_{conv} of $\sim 10\text{-}20 \text{ nm}$ the temperature variation of which is drawn in Fig. 10b. This estimate of L_{conv} is in reasonable agreement with the theoretical calculations^{10,12,13}.

V. CONCLUSIONS

We have realized submicron antidot arrays in thin single NbSe_3 crystals. The comparison between the transport properties respectively in the patterned and unpatterned parts of the crystal reveals specific features of Ohmic conduction and CDW transport in submicron units.

VI. ACKNOWLEDGEMENTS

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Table of sample parameters

Sample	Hole parameters		Spacing		Patterned zone		Unpatterned zone	
	μm		μm		μm		μm	
	D_b	D_c	S_b	S_c	Length	Width	Length	Width
#1	1.5	1.4	3.0	3.7	80	130	540	130
#2	0.3	0.4	0.6	0.6	70	22	620	22

Table gives the dimension of the individual hole and of the periodicity of the antidot array (see Fig. 1) for sample # 1 and # 2. Dimensions of the samples are also indicated.

Figures

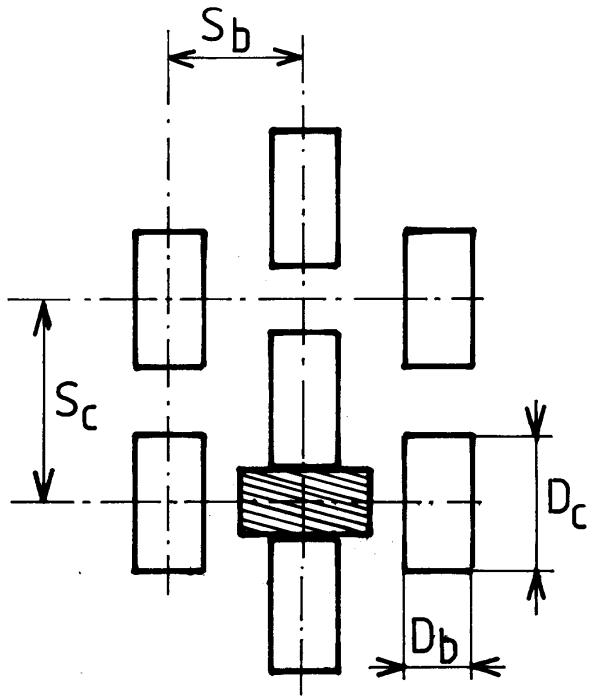


FIG. 1. Schematic picture of the structure with a triangle lattice of holes (antidots). The chain axis is along the X-direction. The projections of holes along the Y-direction overlap in order to block CDW motion in a cell between neighboring holes. Elementary CDW conducting unit is shown as the shaded rectangle

FIG. 2. Scanning electron micrographs of the patterned parts of NbSe_3 . a) sample # 1 on a silicon substrate, b) sample # 2 on a sapphire substrate.

FIG. 3. The geometry of the experiment.

FIG. 4. Temperature dependences of the resistance of NbSe_3 (sample # 2) a) before and after processing (baking the PMMA layer at 160°C), b) of the patterned and unpatterned part after processing.

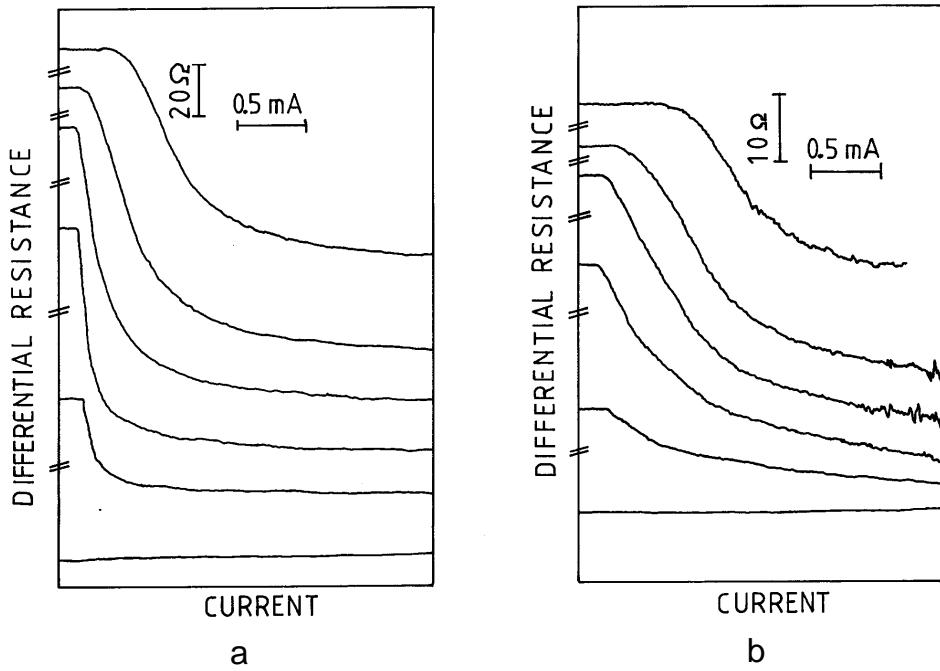


FIG. 5. Variation of the differential resistance $\frac{dV}{dT}$, as a function of current, I , for the patterned and the non-patterned parts of NbSe₃ (sample # 2). The temperatures from top to bottom are as follows : 38.3 K, 42.1, 49.8, 56.9, 62.7, 67.7 K. The corresponding linear resistances (in Ω) are as follows : 105, 106, 118, 107, 87, 76, (for the patterned part) and 118, 141, 151, 139, 98, 66 (for the non-patterned part).

FIG. 6. Temperature dependences of the average threshold current $\langle I_T \rangle$ for initiating the CDW conduction in NbSe₃ (sample # 2).

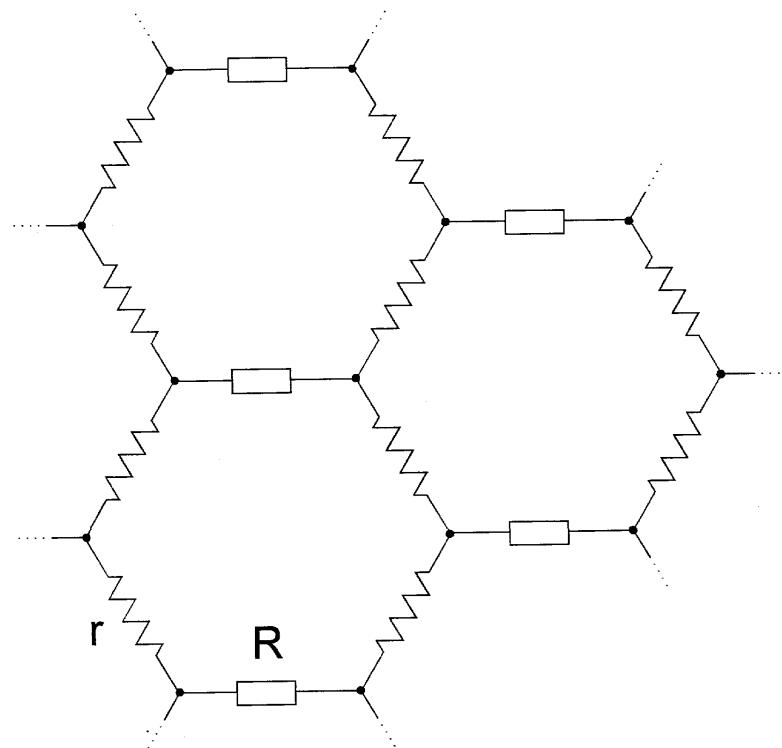


FIG. 7. The equivalent scheme of the patterned part.

FIG. 9. Temperature dependence of the ratio, α , between average threshold current $\langle I_T \rangle$ and $\langle I_T^0 \rangle$ for the patterned and unpatterned parts of NbSe₃ (sample # 1 and # 2).

FIG. 10. a) temperature dependence of the ratio of the CDW conduction in the submicron unit (shaded rectangle in Fig.1) and in the equivalent bulk part for applied current 10 times the average threshold value for NbSe₃ (sample # 2); b) temperature dependence of the conversion length L_{conv} for normal carriers into CDW current (sample # 2). For the procedure to extract L_{conv} from data, see text.